Romanian Reports in Physics, Vol. 65, No. 1, P. 193–203, 2013

M-POINT PHONON EIGENVECTORS OF GRAPHENE OBTAINED BY GROUP PROJECTORS

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Received May 8, 2012

Abstract. We used Wigner's method of group projectors in order to find the phonon eigenvectors of the honeycomb lattice at the M-point of the first Brillouin zone. Our results are in agreement with ones obtained by other methods thus providing an example of an eigenvalue problem solved entirely by symmetry.

Key words: graphene, group theory, phonons.

1. INTRODUCTION

The physics on the honeycomb lattice gain much interest since graphene has been synthesized for the first time in 2004 [1]. This discovery opened different possibilities for both fundamental research and applications. Vibrations of nuclei (phonons), are essential for understanding physical properties of this material. Phonon energies and displacement patterns are a dynamical problem solution. Phonon energies correspond to eigenvalues of the dynamical matrix and displacement patterns can be obtained from eigenvectors of this matrix. It turns out that symmetry of the system can significantly simplify a dynamical problem. The usual consequence of the underlying symmetry of a system, is to reduce the dynamical matrix to a block diagonal form, thus reducing one "large" problem to several "smaller" problems [2]. In the case of the honeycomb lattice the problem of finding displacement patterns of nuclei, for phonons belonging to Γ, M and K points of the first Brillouin zone (BZ), can be completely solved through this simplification. The band structure of graphene has a saddle point at the M-point of the BZ. Such point occurs when the curvature of bands has an opposite sign in two orthogonal directions. This results in a divergence in the density of states and it is well known as the van Hove singularity. The Fermi level in graphene can be tuned over a wide energy region by a chemical doping [3–5] or an electrostatic gating [6–8]. If the deformation of the Fermi surface is induced in graphene, the deformation is the highest in the K-M direction of BZ, and reaches the maximum

when the Fermi level is very close to the van Hove singularity at the M-point. Fundamental interactions between charge, spin and lattice depend very much on the topology of electronic bands, especially in two dimensions when the Fermi level is near the saddle point. Objective of our paper is to present the procedure and results of the honeycomb lattice displacement pattern calculation, for the phonons belonging to the M-point of the first BZ, applying only the symmetry rules, *i.e.* by the method of group projectors.

2. THEORY

A dynamical matrix, whose eigenvalue problem we seek to solve in order to obtain phonon energies and displacement patterns, in general case reads [9]:

$$D_{\alpha\beta}\begin{pmatrix}\mathbf{q}\\ j;l\end{pmatrix}=\frac{1}{\sqrt{M_j M_l}}\sum_{\mathbf{R}}\Phi_{\alpha\beta}\begin{pmatrix}0;\mathbf{R}\\ j;l\end{pmatrix}\exp\left(\mathrm{i}\mathbf{q}\cdot\mathbf{R}\right). \tag{2.1}$$

Here $\mathbf{q}$ is a wave vector in the first BZ, s is a total number of nuclei in the primitive cell, M_j is the mass of j-th nucleus, $\mathbf{R}$ runs over direct lattice, j and l denote the nuclei in the primitive cell, α and β take values x, y or z in the rectangular coordinate system and $\Phi_{\alpha\beta}\begin{pmatrix}\mathbf{0};\mathbf{R}\\ j,l\end{pmatrix}$ are the force constants (*i.e.* second order derivatives of potential energy of nuclei with respect to displacements of nuclei from the equilibrium position). The dimension of the dynamical matrix is equal to $3s$, and its eigenvalue problem is:

$$\sum_{l;\beta}D_{\alpha\beta}\begin{pmatrix}\mathbf{q}\\ j;l\end{pmatrix}e_\beta(l|\mathbf{q})=\omega^2(\mathbf{q})e_\alpha(j|\mathbf{q}). \tag{2.2}$$

Here ω^2 are real, non negative values, with ω being angular frequency of normal mode and displacement of j-th nucleus in $\mathbf{R}$-th primitive cell along axis α is:

$$u_\alpha\begin{pmatrix}\mathbf{R}\\ j\end{pmatrix}=(\sqrt{NM_j})^{-1}\,\mathrm{Re}\left\{e_\alpha(j|\mathbf{q})\exp\{-\mathrm{i}[\mathbf{q}\cdot\mathbf{R}-\omega(\mathbf{q})t]\}\right\}, \tag{2.3}$$

where N is number of primitive cells in the crystal and $e_\alpha(j|\mathbf{q})$ are (in general complex) the components of an eigenvector. To simplify calculations, we have included the phase factors $\exp\left(\mathrm{i}\mathbf{q}\cdot\mathbf{r}_j\right)$ into the phonon eigenvectors, *i.e.* we have

made the substitution $\exp(\mathrm{i}\mathbf{q}\cdot\mathbf{r}_j)e_\alpha(j|\mathbf{q}) \to e_\alpha(j|\mathbf{q})$, where $\mathbf{r}_j$ is an equilibrium position of the j^{th} nucleus. Symmetry of a crystal can help us in solving the equation (2.2) in the following way [2]. For a crystal that belongs to symmorphic space group $T \wedge \underline{G}$ (here we consider only the symmorphic space groups since the honeycomb lattice belongs to such a group), the first term in the product is a group of pure translations and the second is one of crystallographic point groups, for each wave vector $\mathbf{q}$ the group of the wave vector is defined as a subset of operations in $\underline{G}$ that transform this wave vector to its equivalent [2,10]:

$$\underline{G}_{\mathbf{q}} = \left\{\mathbf{g} \in \underline{G} \,|\, \mathbf{g}\mathbf{q} - \mathbf{q} \in \text{InverseLattice}\right\}. \tag{2.4}$$

Consider now an element $\mathbf{g}$ of the group of the wave vector $\mathbf{q}$. It transforms an arbitrary nucleus at $\mathbf{r}_j$ in the primitive cell at the origin to some position (not necessary in the same cell). Accordingly:

$$\mathbf{g}\mathbf{r}_j = \mathbf{r}_l + \mathbf{R}, \tag{2.5}$$

so that $\mathbf{r}_l$ is from the same primitive cell and $\mathbf{R}$ is a vector from direct lattice. l and $\mathbf{R}$ are then functions of j and $\mathbf{g}$. Since the cell we have chosen is primitive, l and $\mathbf{R}$ in the equation (2.5) are uniquely defined. For every wave vector $\mathbf{q}$ and for every element $\mathbf{g}$ of $\underline{G}_{\mathbf{q}}$, the matrix of dynamical representation is:

$$\mathbf{D}_d(\mathbf{g}) = \sum_{j=1}^{s} \exp\left[i\mathbf{q}\cdot\left(\mathbf{g}\mathbf{r}_j - \mathbf{r}_l\right)\right]\mathbf{E}_{lj} \otimes \mathbf{g}. \tag{2.6}$$

Here $\mathbf{g}$ at the right hand side is polar vector representation of $\underline{G}_{\mathbf{q}}$. It maps three dimensional, real vector space into itself. If $\{\mathbf{e}_1, \mathbf{e}_2, \mathbf{e}_3\}$ are the base vectors in this space, then matrix of polar vector representation is given by $\mathbf{g}\mathbf{e}_p = \sum_{m=1}^{3} [\mathbf{g}]_{mp}\, \mathbf{e}_m$ for every p from 1 to 3. $\mathbf{E}_{lj}$ is the s-dimensional square matrix with the element lj equal to one and all other elements equal to zero, whereas $\otimes$ denotes the tensor product. For every element of the group of the wave vector $\mathbf{q}$, matrix $\mathbf{D}_d(\mathbf{g})$ commutes with the dynamical matrix $\mathbf{D}(\mathbf{q})$ given in the formula (2.1). This allows us to proceed in a way that is usual for applying symmetry: we reduce the dynamical representation given by formula (2.6) into irreducible components and

find a symmetry adapted basis in which the dynamical representation and consequently, the dynamical matrix, are reduced to block diagonal forms. In the case of the honeycomb lattice and the wave vector at the M-point every non-equivalent irreducible representation appears in the decomposition of dynamical representation at most once so that the dynamical matrix is diagonalized already in the symmetry adapted basis. This means that symmetry adapted basis vectors are at the same time eigenvectors of the dynamical matrix. In the next section we apply this method to the honeycomb lattice.

3. RESULTS AND DISCUSSION

The honeycomb lattice and the primitive cell we have chosen are shown in the Fig. 1. The lattice consists of regular hexagons with common edges, and the primitive cell contains two nuclei. Apart from the translational symmetry, which is generated by the primitive vectors $\boldsymbol{a}_1$ and $\boldsymbol{a}_2$, the honeycomb lattice has 24 more symmetry elements.

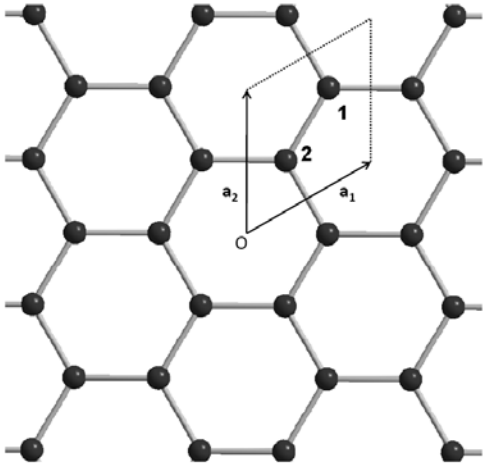

Fig.1 – The honeycomb lattice and the primitive cell that we have chosen.

Those elements (shown in Fig. 2) form point group $\underline{D}_{6h}$. The total symmetry group is a semi direct product of the two and is denoted by Dg80 [11]: $Dg80 = T \wedge \underline{D}_{6h}$.

The reciprocal space and the first BZ for the honeycomb lattice are shown in Fig. 3. The symmetry elements of the reciprocal and direct lattice are identical.

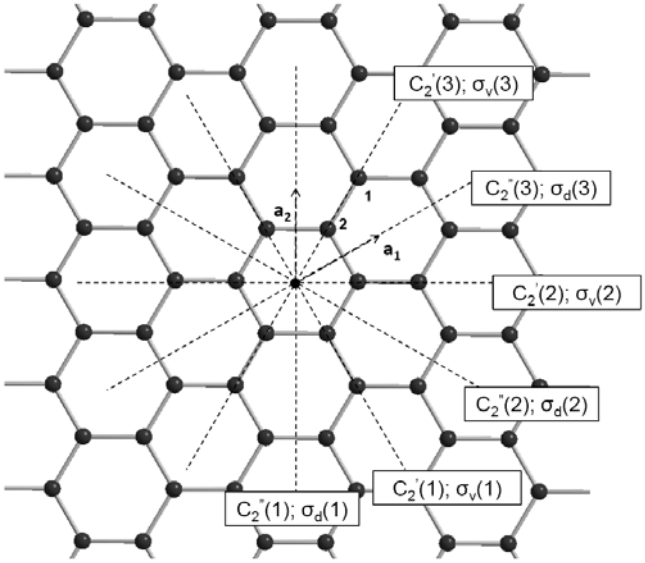

Fig. 2 – Symmetry elements of the honeycomb lattice. Dashed lines denote axes of order 2 and intersections of vertical reflection planes with the horizontal plane. Vertical axis of order 6, inversion center and horizontal reflection plane are also symmetry elements (not shown).

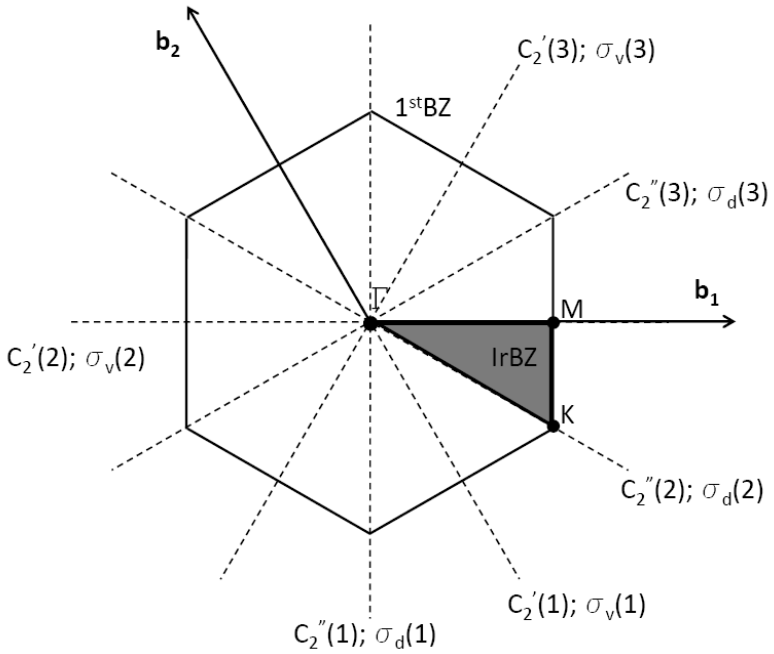

Fig. 3 – The first Brillouin zone of the honeycomb lattice. IrBZ is irreducible part of the first Brillouin zone. Notice M-point at $(1/2)\cdot\mathbf{b}_1$.

The group of the wave vector at the point M is $\underline{D}_{2h}$ and consists of the following elements: identity $\hat{E}$; three mutually orthogonal axes of order two $\hat{C}_2(z)$, $\hat{C}_2'(2)$, $\hat{C}_2''(1)$; inversion $\hat{i}$; and three reflection planes $\hat{\sigma}_h$, $\hat{\sigma}_d(1)$, $\hat{\sigma}_v(2)$. The matrices of the dynamical representation obtained from the formula (2.6) are given in the Table 1. The basis vectors according to which matrices of the polar vector representation which enter in (2.6) were calculated are given in the Fig. 4. As an example we calculate the matrix $\mathbf{D}_d\left(\hat{C}_2''(1)\right)$.

$$\begin{aligned} \hat{C}_2''(1)\mathbf{r}_1 &= \mathbf{r}_2 - \boldsymbol{a}_1 + \boldsymbol{a}_2 \\ \hat{C}_2''(1)\mathbf{r}_2 &= \mathbf{r}_1 - \boldsymbol{a}_1 \end{aligned}, \tag{3.1}$$

since
$$\begin{aligned} \exp\left[\mathrm{i}\mathbf{q}_M \cdot (-\boldsymbol{a}_1 + \boldsymbol{a}_2)\right] &= \exp\left[(\mathrm{i}/2)\mathbf{b}_1 \cdot (-\boldsymbol{a}_1 + \boldsymbol{a}_2)\right] = \exp\left(-\mathrm{i}\pi\right) = -1 \\ \exp\left[\mathrm{i}\mathbf{q}_M \cdot (-\boldsymbol{a}_1)\right] &= \exp\left[(\mathrm{i}/2)\mathbf{b}_1 \cdot (-\boldsymbol{a}_1)\right] = \exp(-\mathrm{i}\pi) = -1 \end{aligned} \tag{3.2}$$

to the element $\hat{C}_2''(1)$ corresponds the matrix

$$\begin{pmatrix} \hat{0} & -\hat{C}_2''(1) \\ -\hat{C}_2''(1) & \hat{0} \end{pmatrix}.$$

Table 1

Matrices of the dynamical representation obtained using formula 2.6.
Adopted coordinate system is given in Fig. 4

g	$\hat{E}$	$\hat{C}_2(z)$	$\hat{i}$	$\hat{\sigma}_h$
$\mathbf{D}_d(\mathbf{g})$	$\begin{pmatrix} 1&0&0&0&0&0\\ 0&1&0&0&0&0\\ 0&0&1&0&0&0\\ 0&0&0&1&0&0\\ 0&0&0&0&1&0\\ 0&0&0&0&0&1 \end{pmatrix}$	$\begin{pmatrix} 0&0&0&1&0&0\\ 0&0&0&0&1&0\\ 0&0&0&0&0&-1\\ 1&0&0&0&0&0\\ 0&1&0&0&0&0\\ 0&0&-1&0&0&0 \end{pmatrix}$	$\begin{pmatrix} 0&0&0&1&0&0\\ 0&0&0&0&1&0\\ 0&0&0&0&0&1\\ 1&0&0&0&0&0\\ 0&1&0&0&0&0\\ 0&0&1&0&0&0 \end{pmatrix}$	$\begin{pmatrix} 1&0&0&0&0&0\\ 0&1&0&0&0&0\\ 0&0&-1&0&0&0\\ 0&0&0&1&0&0\\ 0&0&0&0&1&0\\ 0&0&0&0&0&-1 \end{pmatrix}$
g	$\hat{C}_2'(2)$	$\hat{C}_2''(1)$	$\hat{\sigma}_d(1)$	$\hat{\sigma}_v(2)$
$\mathbf{D}_d(\mathbf{g})$	$\begin{pmatrix} -\frac{1}{2}&\frac{\sqrt{3}}{2}&0&0&0&0\\ \frac{\sqrt{3}}{2}&\frac{1}{2}&0&0&0&0\\ 0&0&-1&0&0&0\\ 0&0&0&-\frac{1}{2}&\frac{\sqrt{3}}{2}&0\\ 0&0&0&\frac{\sqrt{3}}{2}&\frac{1}{2}&0\\ 0&0&0&0&0&-1 \end{pmatrix}$	$\begin{pmatrix} 0&0&0&-\frac{1}{2}&\frac{\sqrt{3}}{2}&0\\ 0&0&0&\frac{\sqrt{3}}{2}&\frac{1}{2}&0\\ 0&0&0&0&0&1\\ -\frac{1}{2}&\frac{\sqrt{3}}{2}&0&0&0&0\\ \frac{\sqrt{3}}{2}&\frac{1}{2}&0&0&0&0\\ 0&0&1&0&0&0 \end{pmatrix}$	$\begin{pmatrix} 0&0&0&-\frac{1}{2}&\frac{\sqrt{3}}{2}&0\\ 0&0&0&\frac{\sqrt{3}}{2}&\frac{1}{2}&0\\ 0&0&0&0&0&-1\\ -\frac{1}{2}&\frac{\sqrt{3}}{2}&0&0&0&0\\ \frac{\sqrt{3}}{2}&\frac{1}{2}&0&0&0&0\\ 0&0&-1&0&0&0 \end{pmatrix}$	$\begin{pmatrix} -\frac{1}{2}&\frac{\sqrt{3}}{2}&0&0&0&0\\ \frac{\sqrt{3}}{2}&\frac{1}{2}&0&0&0&0\\ 0&0&1&0&0&0\\ 0&0&0&-\frac{1}{2}&\frac{\sqrt{3}}{2}&0\\ 0&0&0&\frac{\sqrt{3}}{2}&\frac{1}{2}&0\\ 0&0&0&0&0&1 \end{pmatrix}$

Here $\hat{0}$ is a square, three dimensional zero matrix. By representing $\hat{C}_2''(1)$ in the basis given in the Fig. 4 we obtain the corresponding matrix in the Table 1. From

the Table 1 we can find the characters of the dynamical representation for the point *M* in the first BZ and use them to decompose this representation. The table of non equivalent irreducible representations for the point group $\underline{D}_{2h}$ necessary for this step, is adopted from [12]. The dynamical representation decomposition for point *M* is the following:

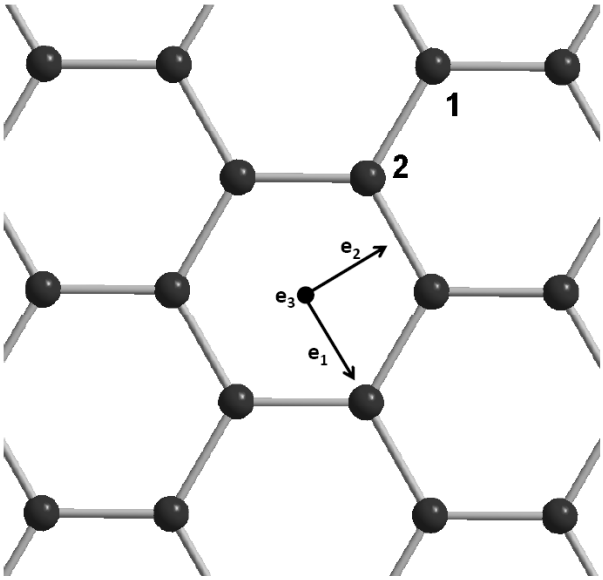

Fig. 4 – Coordinate system for construction of dynamical represetation matrices. Vector $\mathbf{e_3}$ is perpendicular to the drawing plane and points towards the reader.

$$MA_g + MB_{1g} + MB_{3g} + MB_{1u} + MB_{2u} + MB_{3u}\,. \tag{3.3}$$

Here for instance, MA_g denotes a non equivalent irreducible representation of the diperiodic group Dg80 obtained by an induction from the M-point wave vector and the non equivalent irreducible representation A_g of the group of this wave vector. Since the group of the *M*-point wave vector and the point group of the crystal have 8 and 24 elements, respectively, it follows that such irreducible representations of Dg80 are 3 dimensional. This is in accordance with the work of I. Milošević *et al.* where all non equivalent irreducible representations of generators of all 80 diperiodic groups were constructed [13]. From (3.3) it follows that each irreducible representation in M-point phonon classification appears at most once. This allows us to find phonon eigenvectors using group projectors. The formula for group projector that corresponds to the non equivalent irreducible representation α of the group $\underline{G}$ is [2]:

$$\hat{P}_{\alpha} = \frac{1}{|\underline{G}|} \sum_{\mathbf{g} \in \underline{G}} \chi_{\alpha}^{*}(\mathbf{g}) \mathbf{D}_{d}(\mathbf{g}) \,. \tag{3.4}$$

Here $|\underline{G}|$ is the order of the group $\underline{G}$ and χ_{α} is the character of a non equivalent irreducible representation α of the group $\underline{G}$. In our case all non equivalent irreducible representations are one dimensional so the characters and the irreducible representations are identical. Group projectors are shown in the Table 2.

Table 2

Group projectors obtained using formula 3.4. Fig. 4 shows adopted coordinate system

α	A_g	B_{1g}	B_{3g}
$8\hat{P}_{\alpha}$	$\begin{pmatrix} 1 & \sqrt{3} & 0 & 1 & \sqrt{3} & 0 \\ \sqrt{3} & 3 & 0 & \sqrt{3} & 3 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 1 & \sqrt{3} & 0 & 1 & \sqrt{3} & 0 \\ \sqrt{3} & 3 & 0 & \sqrt{3} & 3 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix}$	$\begin{pmatrix} 3 & -\sqrt{3} & 0 & 3 & -\sqrt{3} & 0 \\ -\sqrt{3} & 1 & 0 & -\sqrt{3} & 1 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 3 & -\sqrt{3} & 0 & 3 & -\sqrt{3} & 0 \\ -\sqrt{3} & 1 & 0 & -\sqrt{3} & 1 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix}$	$\begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 4 & 0 & 0 & 4 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 4 & 0 & 0 & 4 \end{pmatrix}$
α	B_{1u}	B_{2u}	B_{3u}
$8\hat{P}_{\alpha}$	$\begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 4 & 0 & 0 & -4 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & -4 & 0 & 0 & 4 \end{pmatrix}$	$\begin{pmatrix} 1 & \sqrt{3} & 0 & -1 & -\sqrt{3} & 0 \\ \sqrt{3} & 3 & 0 & -\sqrt{3} & -3 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ -1 & -\sqrt{3} & 0 & 1 & \sqrt{3} & 0 \\ -\sqrt{3} & -3 & 0 & \sqrt{3} & 3 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix}$	$\begin{pmatrix} 3 & -\sqrt{3} & 0 & -3 & \sqrt{3} & 0 \\ -\sqrt{3} & 1 & 0 & \sqrt{3} & -1 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ -3 & \sqrt{3} & 0 & 3 & -\sqrt{3} & 0 \\ \sqrt{3} & -1 & 0 & -\sqrt{3} & 1 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix}$

The group projector for an irreducible representation α is a linear operator that maps an arbitrary vector from vector space into projection of that vector onto the subspace which corresponds to α. As an example, we use projection on MA_g (a, b, c, d, f, g are arbitrary real numbers):

$$\hat{P}_{MA_g} \begin{pmatrix} a \\ b \\ c \\ d \\ f \\ g \end{pmatrix} = \frac{1}{8}(a + \sqrt{3}b + d + \sqrt{3}f) \begin{pmatrix} 1 \\ \sqrt{3} \\ 0 \\ 1 \\ \sqrt{3} \\ 0 \end{pmatrix} = \frac{1}{4}(a + \sqrt{3}b + d + \sqrt{3}f) \begin{pmatrix} 1/2 \\ \sqrt{3}/2 \\ 0 \\ 1/2 \\ \sqrt{3}/2 \\ 0 \end{pmatrix}$$

so that the subspace corresponding to MA_g is spaned by the vector $\begin{pmatrix} 1/2 \\ \sqrt{3}/2 \\ 0 \\ 1/2 \\ \sqrt{3}/2 \\ 0 \end{pmatrix}$.

The components of this vector are displacements of nuclei 1 and 2 in the primitive unit cell shown in Fig. 1, represented in the basis shown in Fig. 4 (first number is u_{1x}, second is u_{1y} etc.). Displacements of other nuclei in the crystal can be calculated with a help of the formula (2.3). The results of calculations using the group projectors for all non equivalent irreducible representations that appear in the decomposition (3.3) are given in Fig. 5. Assignation of normal modes is consistent with the symmetry system we assumed. One can be convinced that the obtained results are correct by comparing with results obtained in the literature by other methods. M. Mohr *et al.* [14] have calculated eigenvectors and energies of normal modes by fitting the force constants to observed energies. Their modes obtained numerically are identical to ours. The differences in the symmetry assignments are due to permutation between the symmetry elements. Our results are also in agreement with those obtained by a molecular based approach published in [15], although their method allows a determination of the in-plane phonons only. We see that the modes MA_g and MB_{2u} are longitudinal, while MB_{1g}, MB_{3g}, MB_{1u} and MB_{3u} are transverse. Since non equivalent irreducible representations of Dg80 belonging to the *M*-point are three dimensional, two more modes with the same energy correspond to each phonon mode presented in Fig. 5. This fact is described by the formula $(\forall \mathbf{g} \in \underline{G})\,\omega(\mathbf{gq}) = \omega(\mathbf{q})$, which is valid for every $\mathbf{q}$ in the reciprocal space. One can obtain the displacement patterns of these modes by rotating a corresponding part of Fig. 5 by ± 60 degrees. One cannot obtain phonon energies using symmetry argument only, but could easily find them from dynamical matrix because its eigenvectors are already known. In our case the dynamical matrix for the point *M* can be completely reconstructed from phonon energies. That is not the case for a dynamical matrix that has eigenvectors not completely determinable from the symmetry, where knowledge of both eigenvalues and eigenvectors is necessary for determining the dynamical matrix. Since eigenvectors are not measurable quantities (except if they are completely determined by symmetry) problem of fitting force constants to observed energies has not a unique solution. As we mentioned earlier, the procedure we used, can be applied to the Γ and K

points also, but for the Γ point it is easier to find eigenvectors by dividing modes to in- and out-of-plane modes and subtract movements that belong to acoustic phonons [16].

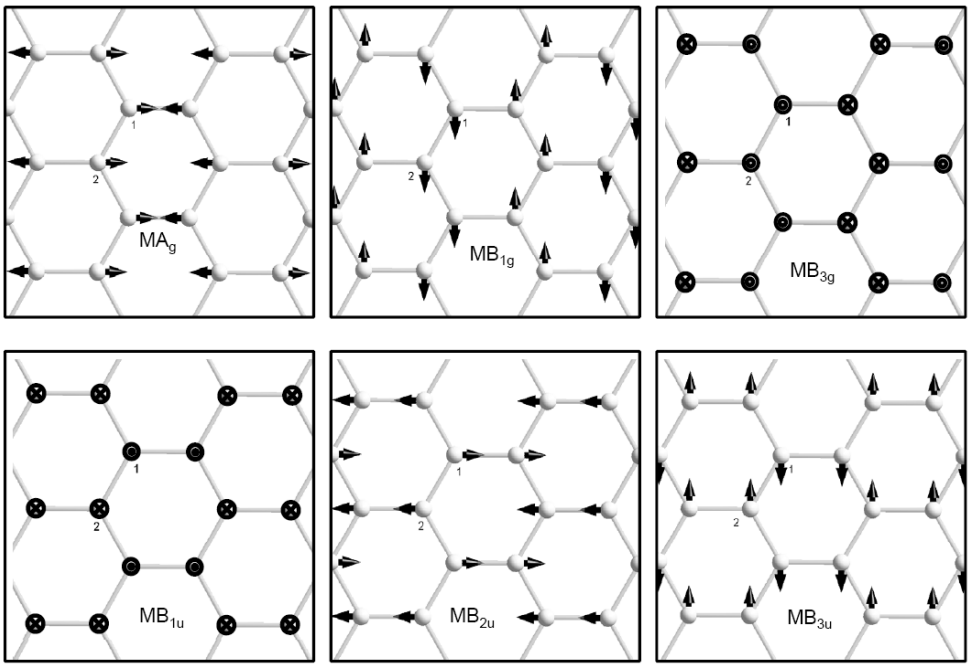

Fig. 5 – M-point phonon eigenvectors of graphene obtained by group projectors.

4. CONCLUSIONS

It is useful to seek for a solution of a physical problem in more than one way, especially to search for an elegant solution. Application of symmetry mostly simplifies solving dynamical problem, but sometimes it completely solves the problem as in the case of phonon eigenvectors in the high symmetry points of graphene's BZ. In this paper, we have presented complete method of group projectors used to classify *M*-point phonons in a crystal belonging to the graphene honeycomb lattice and to find their eigenvectors. Eigenvectors found by this simpler procedure are in accordance with the results obtained by other, more conventional methods done for graphene.

Acknowledgments. This work was supported by Serbian Ministry of Education and Science under the project numbers OI 171005 and III 45016, and by European Union under the project EU FP7 NIM_NIL.

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